<Final Report>

Surface-Templated Assembly for Mass-Production of ZnO Nanowire-Based Integrated Devices

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Abstract

Recently, nanowire-based devices draw large attention as next-generation device architecture, while a lack of mass-production method for such devices has been holding back their practical applications. As proposed in our original proposal, we developed a method to mass-produce ZnO nanowire-based integrated devices, where self-assembled monolayer patterns on the substrates direct the assembly and alignment of ZnO nanowires from the solution without relying on any external forces. As a proof of concepts, we assembled and aligned ZnO nanowires on Au and SiO_2 surfaces. Furthermore, we successfully demonstrated *the large-scale fabrication of ZnO nanowire-based circuits.* As far as the PI knows, this is the first successful demonstration for large-scale fabrication of ZnO-based integrated devices. The developed method may pave the way toward ZnO-based device industry in the future.

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14. ABSTRACT
A method to mass-produce ZnO nanowire-based integrated devices has been investigated. The approach is to employ self-assembled monolayer patterns on the substrates, directly assemble and align ZnO nanowires from the solution without relying on any external forces. As a proof of concepts ZnO nanowires were assembled and aligned on Au and SiO2 surfaces. Furthermore, the large-scale fabrication of ZnO nanowire-based circuits were successfully demonstrated. It is understood that this is the first successful demonstration for large-scale fabrication of ZnO-based integrated devices. The developed method can contribute to ZnO-based device commercial applications in the future.

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1. Introduction

ZnO nanowires can be utilized to build various high-performance nanoelectronic devices such as biosensors, UV nanosensors, nanoscale UV emitters, etc. However, the major bottleneck holding back their practical applications is a lack of a high-throughput manufacturing method for fabrication of such devices. Since ZnO nanowires are first synthesized in a powder form, one has to pick up and assemble individual nanowires onto the substrate to build integrated devices. For this reason, fabrication of ZnO nanowire devices has been an extremely time-consuming process, and, in fact, it is virtually impossible to build integrated devices that often require millions of nanowires.

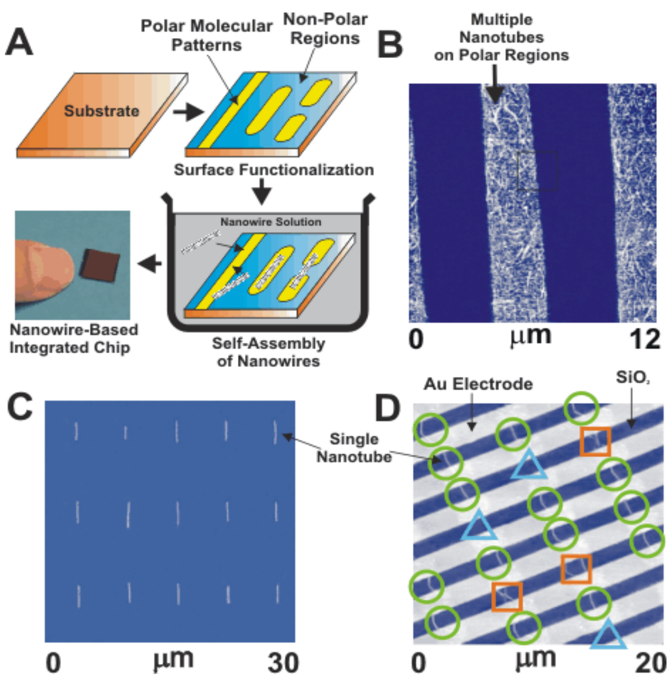

Figure 1. A massive assembly process (A) for nanowire-based devices is utilized to create various carbon nanotube-based structures ((B)-(D)).

Recently, we successfully utilized the *self-assembly* strategy to assemble and align single-walled carbon nanotubes (swCNTs) onto solid substrate for the mass-production of nanotube-based integrated circuits [*Nature* **425**, 36 (2003)]. In this process, specific nanoscale regions on the solid substrate are first functionalized with organic molecular monolayer that can attract nanotubes, and other regions are passivated with inert molecules (Figure 1A). Molecular patterning has been done via dip-pen nanolithography (DPN) and microcontact printing. When the substrate is placed in the nanowire solution, nanowires are attracted toward the specific regions and they assemble to form pre-designed structures. Utilizing this strategy, we successfully assembled millions of carbon nanotube-based circuit structures (Figure 1B-D). Importantly, the same strategy could be applied for other nanowire devices simply by utilizing proper organic molecular.

2. Objectives

The proposed objectives of this project are: 1) the development of a high-throughput manufacturing method based on self-assembly strategy for ZnO nanowire-based integrated device fabrication and 2) the integration of developed method with industrial standard semiconductor processes so that it can be readily utilized for industrial applications.

3. Status of Research Project

As proposed in our original proposal, *we successfully developed the method to mass-produce ZnO-based integrated devices and completely integrated our method with industrial standard semiconductor processes* so that our method is readily accessible to current industry.

The basic steps of the fabrication process for ZnO nanowire devices are depicted in Figure 2. First, negatively-charged and neutral regions are created on solid substrates. Neutral regions on Au and SiO_2 substrates can be generated by depositing 1-octadecanethiol and 1-octadecyltrichlorosilane molecules, respectively. Self-assembled monolayer molecules such as

16-mercaptohexadecanoic acid can be utilized to generate negatively-charged regions on Au. SiO_2 surfaces can be utilized as negatively-charged surfaces without molecular coating because of its natural charges. When the substrate is placed in the ZnO nanowire solution, ZnO nanowires are adsorbed onto negatively-charged region, and they self-align along the patterns. Since the assembled nanowires form stable structures, one can continue additional microfabrication to fabricate electrodes or more complicated circuit structures.

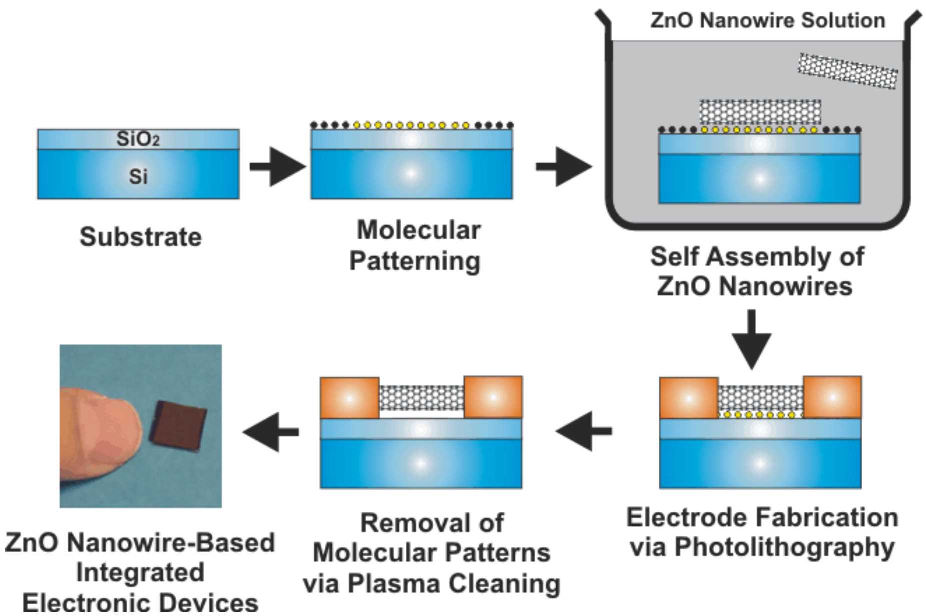

Figure 2. Basic steps for the proposed massive-assembly process for ZnO nanowire-based devices.

A key progress which allows us to integrate this method with conventional semiconductor process is the development of molecular patterning method using industrial standard photolithography. Here, we utilized photoresist patterns as a resist to deposit molecular layers onto specific regions on the substrate. By patterning molecular layers via the standard photolithography method, one can perform the entire steps using only conventional microfabrication facilities, and our method is now readily accessible to current device industry.

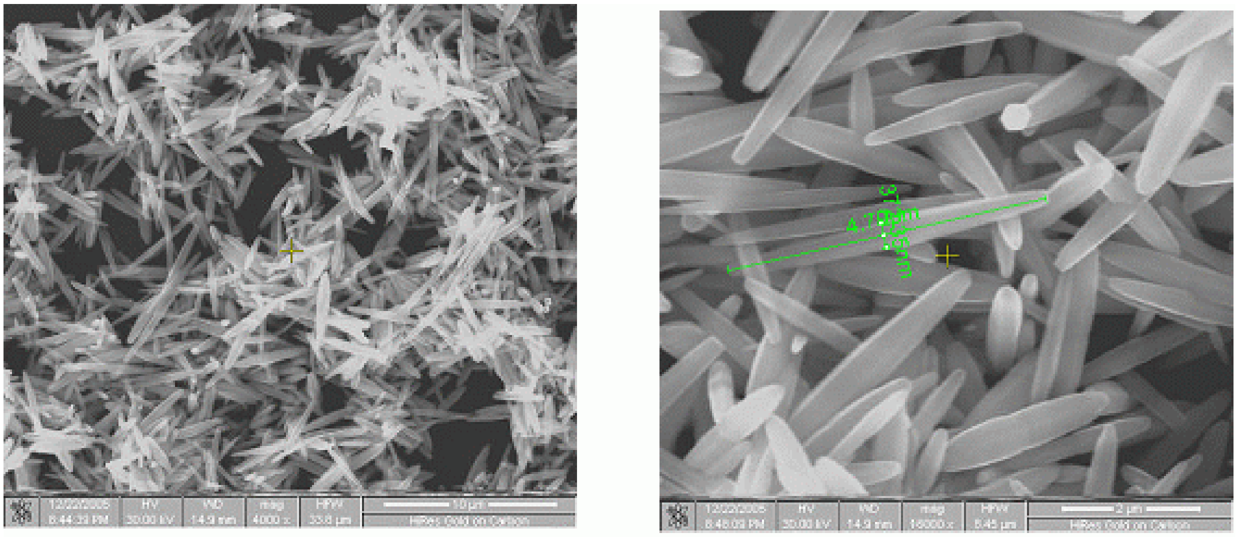

Figure 3. SEM images of as synthesized ZnO nanowires.

ZnO nanowires are synthesized in sol-gel method as reported before [*Adv. Mater.* **15**, 464, (2003)]. The sol-gel method allows us to grow large amount of ZnO nanowires in a relatively short time period. Figure 3 shows the scanning electron microscopy (SEM) images of as-synthesized ZnO nanowires. The nanowires are dispersed in aqueous solution for the assembly process.

When molecule-patterned substrates are placed in the ZnO solution, ZnO NWs are attracted to negatively-charged regions, and they self-align along the regions. Figure 4 shows ZnO nanowires assembled and aligned onto negatively-charged regions on Au and SiO_2. We can see that ZnO nanowires are assembled only onto negatively charged surface regions. Furthermore, they align horizontally to stay inside the negatively-charged regions.

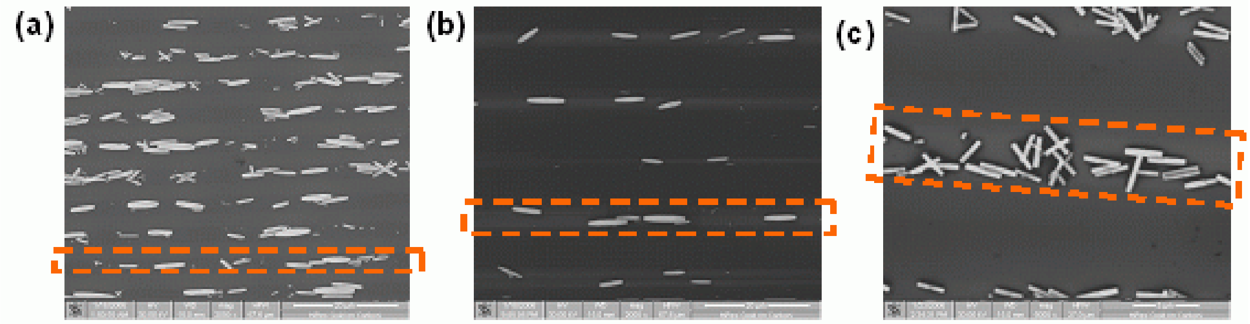

Figure 4. SEM image of ZnO nanowires assembled onto negatively-charged regions (marked by square) on (a) Au and (b),(c) SiO_2 surfaces.

Since the assembled ZnO structures form stable structures, we can continue additional microfabrication processes to add electrodes. Figure 5 shows a ZnO junction and its electric properties. We utilized conventional photolithography and lift-off method to fabricate the nanowire junction. It should be noted that the entire process including molecular patterning has been done using only conventional semiconductor fabrication facilities.

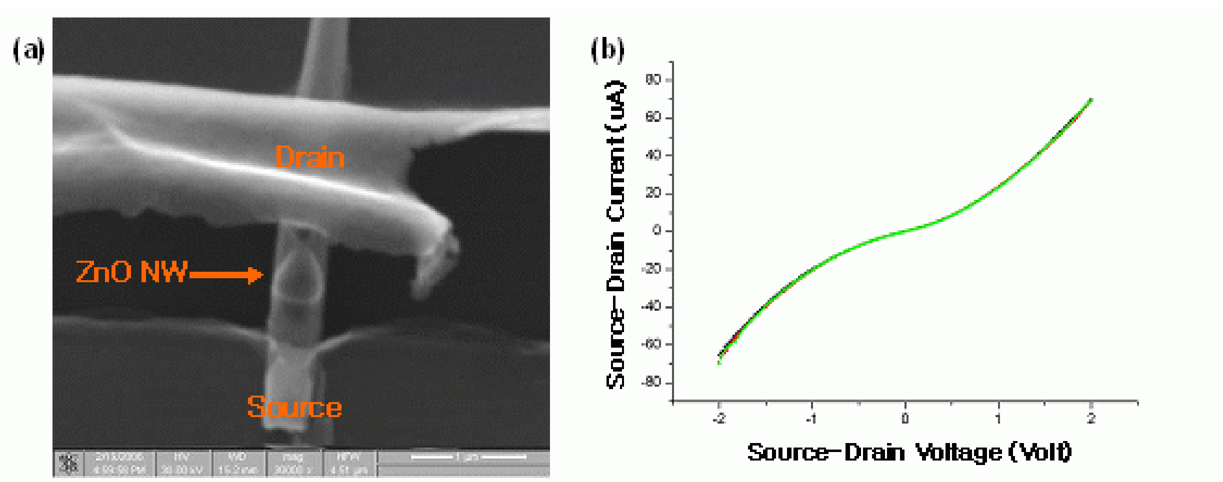

Figure 5. (a) SEM image of ZnO nanowire junction. (b) Electrical measurement result.

The uniqueness and advantages of our approach are: 1) our method does not rely on any external forces such as flow cells, electric or magnetic field etc, and 2) it is compatible with conventional microfabrication processes. Previously assembly methods have to use external forces to precisely control the orientations of individual nanowire devices, which dramatically increases the complication of the fabrication processes. Significantly, since our method does not require any harsh treatment (e.g. high temp) onto the substrate, and they can be readily utilized on various substrates such as glass and even on pre-existing circuit structures.

4. Conclusions

We developed a method to mass-produce ZnO-based integrated devices. As a proof of concepts, we demonstrated the wafer-scale assembly of ZnO nanowires on Au and SiO_2 and the massive fabrication of ZnO-based electrical junctions. The developed method can be utilized to mass-produce general ZnO nanowire-based devices including biological sensors, UV emitters, UV detectors, etc. Considering that the manufacturing issue has been a major problem in nanowire-based device applications, the developed method can be a key bridge connecting from current academic research about ZnO nanowire devices to practical applications. In addition, the developed process may be extended to other metal-oxide nanowires because metal oxide surfaces usually have similar chemical properties.

5. Future Works

Even though our method allows us to mass-produce ZnO nanowire-based device structures, practical applications of such devices still need further research. In the future, we will continue to improve the electric properties of ZnO nanowire-based circuits. Especially, we will try different electrode materials and annealing process to reduce the contact resistance between electrodes and ZnO nanowires. Once the electrical properties of ZnO circuits are optimized, we should be able to explore various practical applications using ZnO NW-based circuits such as biosensors, UV-sensors, etc.